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Molybdenum Coatings on Beam Stopper / Beam Chopper Target

Internal Report

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I. Background

Beams stoppers at Spallation Neutron Source (SNS) are made from TZM (>99% Mo with 0.5% Ti and 0.08% Zr). To reduce radiation caused by nuclear reaction of accelerated protons with titanium, it is highly desirable to have a relatively thick coating of titanium-free molybdenum on the TZM base material. The coating must be well adherent, chemically pure, and uniform.

The general geometry of the beam stop is shown in Figure 1.

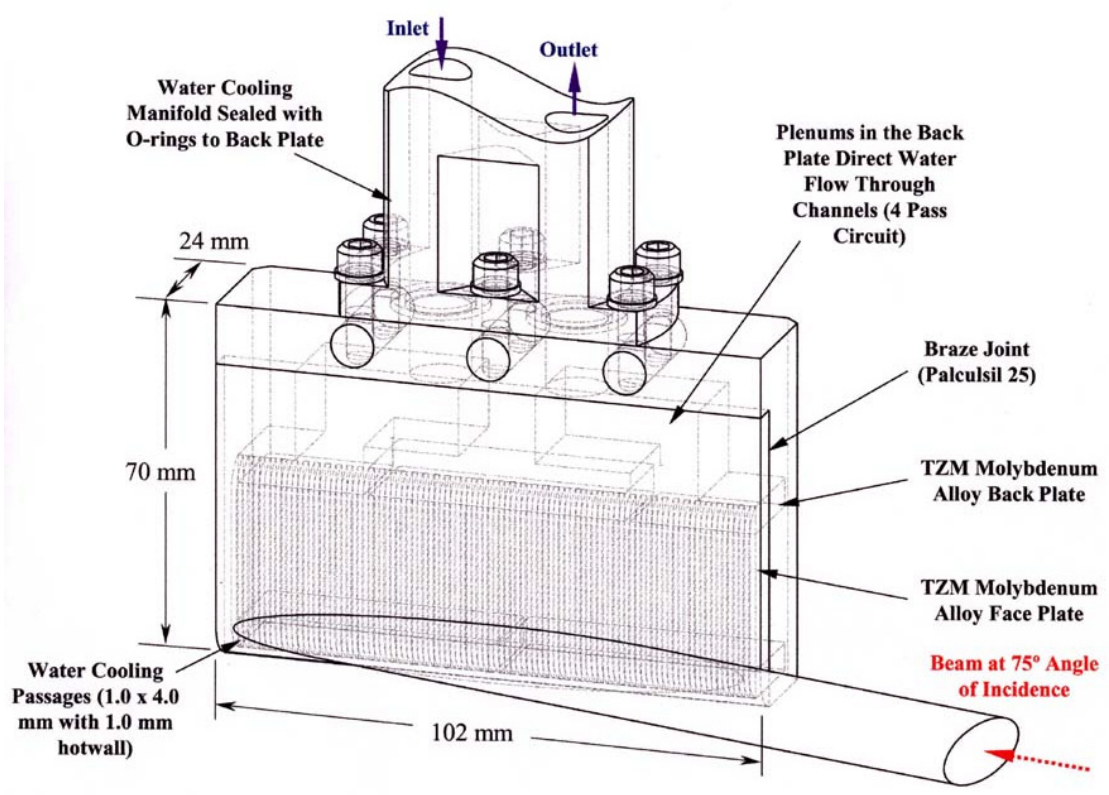

Figure 1: TZM beam stop to be coated with pure molybdenum.

The plasma applications group has developed thin film coating capabilities especially based on pulsed, filtered cathodic arcs. Well adherent films of high quality have been made in this way for several applications. The equipment for pulsed arcs, however, is limited to relatively thin films (much less than one micron), and deposition of reactive metal films tend to have a surprisingly high percentage of oxygen and hydrogen, which was found to be due to the reaction of the film surface atoms with residual water vapor of the process chamber [Schneider, 2000 #1723].

II. Approach

To address the thickness limitation as well as the contamination issue, the arc deposition process must be done in a continuous mode. Based on similar designs published in the literature, we have developed a DC

cathodic arc source suitable for thick molybdenum coatings. In the following, the development of the source is and the deposition of molybdenum films is briefly described.

III. Numerical Simulation

In order to determine the thickness of the coating, one would need the penetration depth of hydrogen ions (protons) for the ion energy of 2.5 MeV with 75° angle to the surface normal. The mean projected range and sputter rate was calculated using the Monte Carlo Code T-DYN 4.0. One obtains 6.95 μm as the projected range and 2×10^{-4} as the sputter yield. The definition of the mean projected range can be seen from Fig. 2.

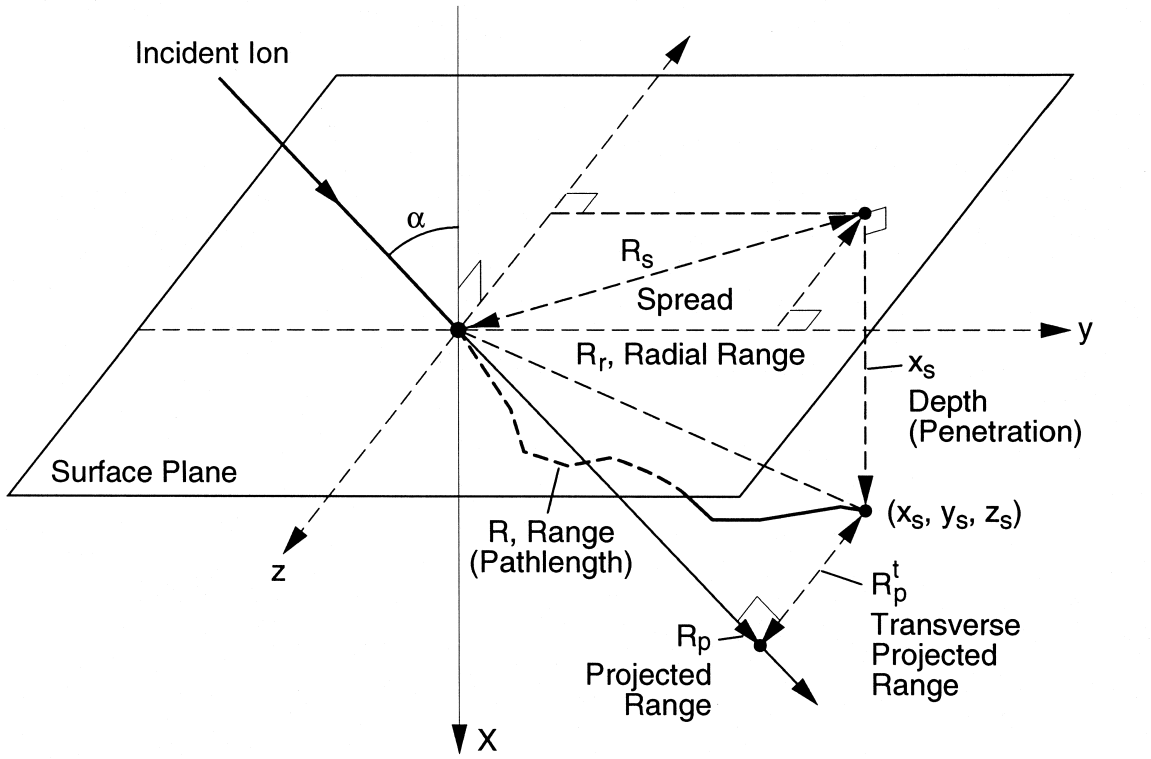

Figure 2. Definition of Projected Range for implanted ions.

The depth can thus easily calculated as $D = R_p \cos\alpha$, which gives $D \approx 1.8\ \mu\text{m}$. One should keep in mind that this depth is an average, and that some ions will penetrate deeper. Another factor is that ions do not loose their energy linearly or supply energy at the location of final rest. Most of the energy is deposited just before the length of the projected range. Yet another factor is that during the lifetime of the beam stopper in operation, some sputtering will occur, gradually reducing the thickness of the molybdenum coating. In conclusion, the coating should be thicker than 2 μm, but does not need to be as thick as 10 μm. We will aim for a thickness in the range 6-10 μm.

IV. Plasma Source Development

For continuous operation, the plasma source must be equipped with a strong magnetic field that facilitates cathode spot motion ("steered arc" [Boxman, 1995 #35]). We used a CoSm magnet with axial magnetization from a previous experiment. This magnet was mounted a preliminary setup (Fig. 3) to test various shapes of cathodes (Fig. 4).

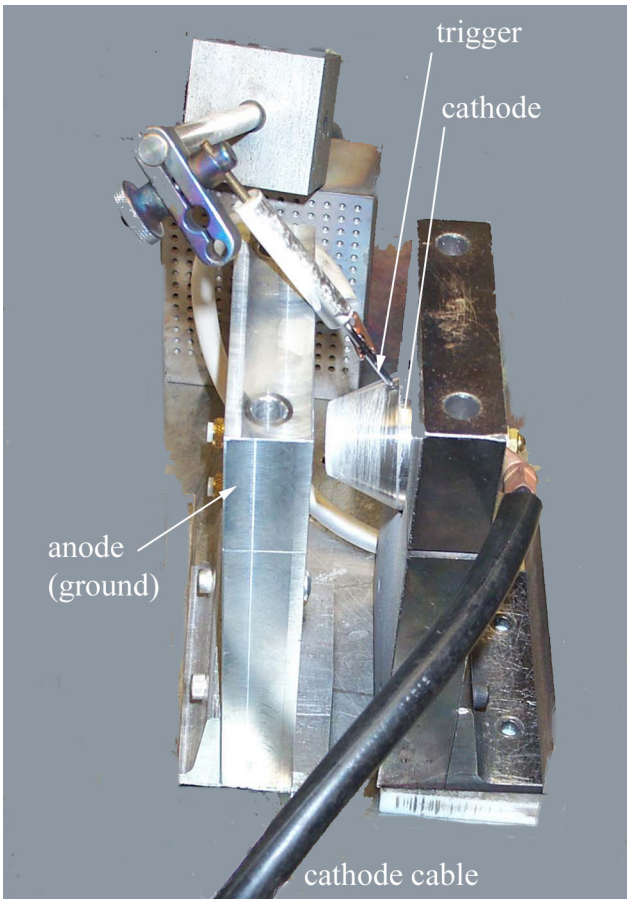

Figure 3: Set-up to develop a suitable plasma source with a strong magnetic field from a permanent magnet, a motion-free trigger. This setup is not water-cooled and was only used for pulses of ~100 ms duration.

Figure 4: Experimental cathodes of various shapes and materials used for the initial source development. The base diameter is 2 inch for all cathodes.

We found suitable conical geometry and made a Mo cathode with that geometry to be used in a water-cooled source. This source was CAD designed; drawings are stored on LBNL server of the Engineering Division. The principal outline of the water-cooled source is shown in Figure 5.

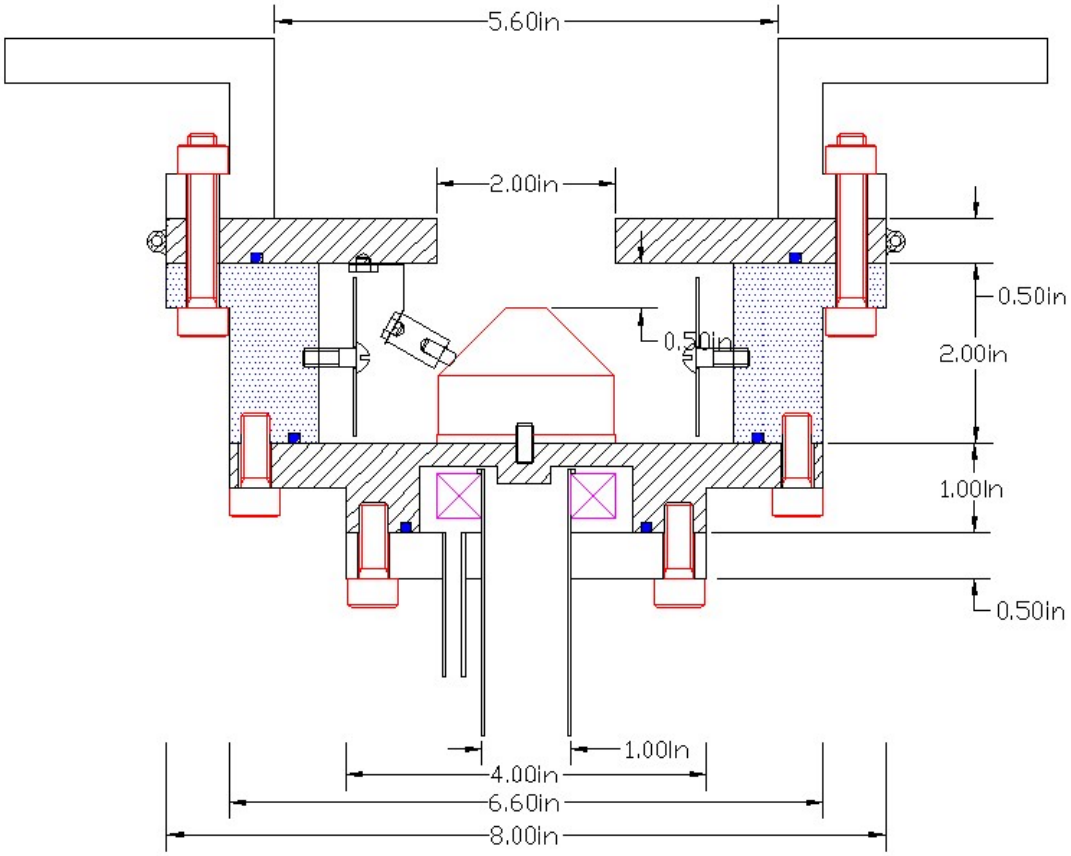

Figure 5: Preliminary outline drawing of DC arc source; the final drawing can be found at LBNL server of the Engineering Division.

The source was manufactured at LBNL. Figures 6-8 show the source from various positions.

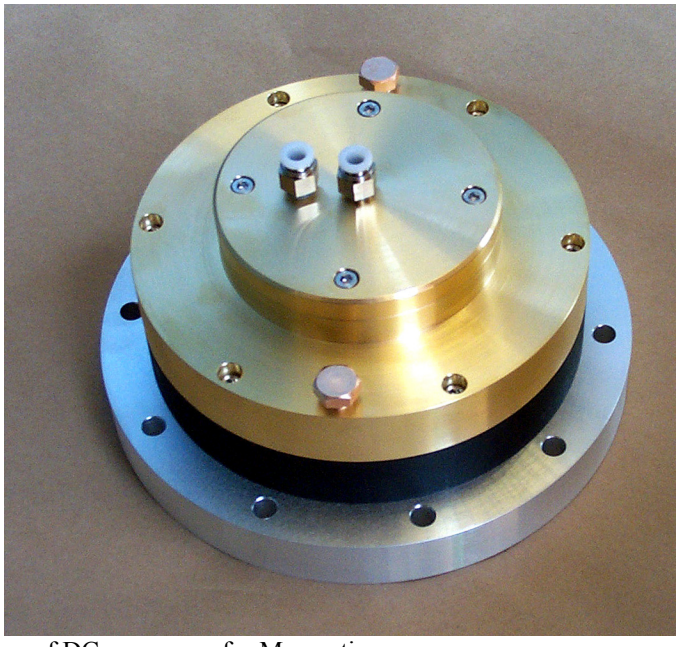

Figure 6. Cathode View of DC arc source for Mo coating.

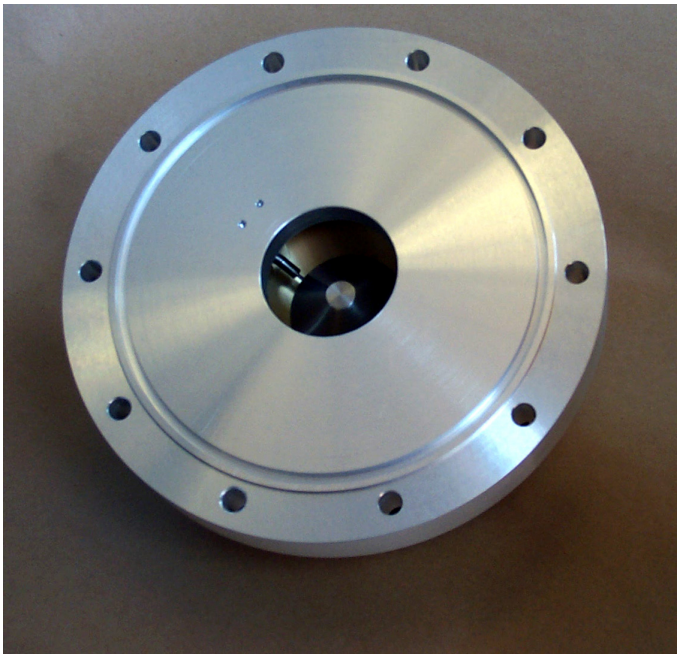

Figure 7. Anode view of the cathodic arc source

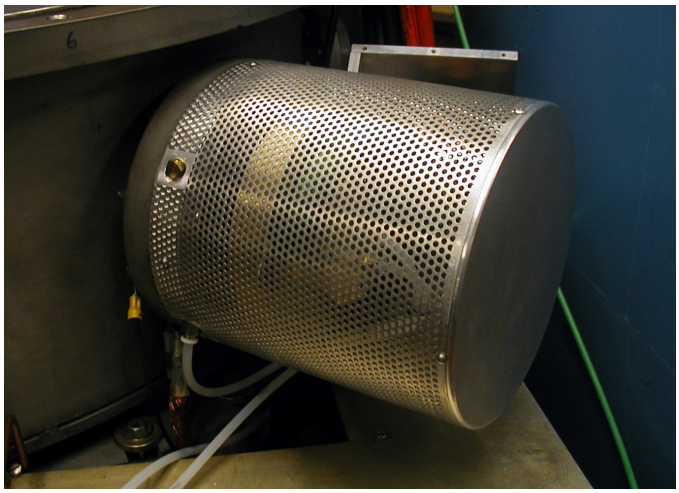

Figure 8: Source mounted to a high vacuum chamber; the perforated part is a safety shield that prevents the operator from touching cathode parts at high negative potential.

V. Initial Test Coatings on TZM Substrates

In order to determine the deposition rate and film adhesion, a series of test coatings have been performed. For these tests we used the same TZM material as used for the real beam stoppers. These tests also served the purpose to verify the reliability of the new source. The arc source was operated with increasing pulse length from an existing DC power supply (max. 200 V, max. 200 A). The power supply was equipped with a custom-made high-current transistor switch unit, allowing the operator to use arc discharges of arbitrary pulse duration. Additionally, the power supply had a booster stage of max. 600 V, as described in Ref. [Anders, 1998 #196], promoting reliable triggering of the arc.

Figure 9 shows how test substrates of TZM were mounted on a water-cooled substrate holder.

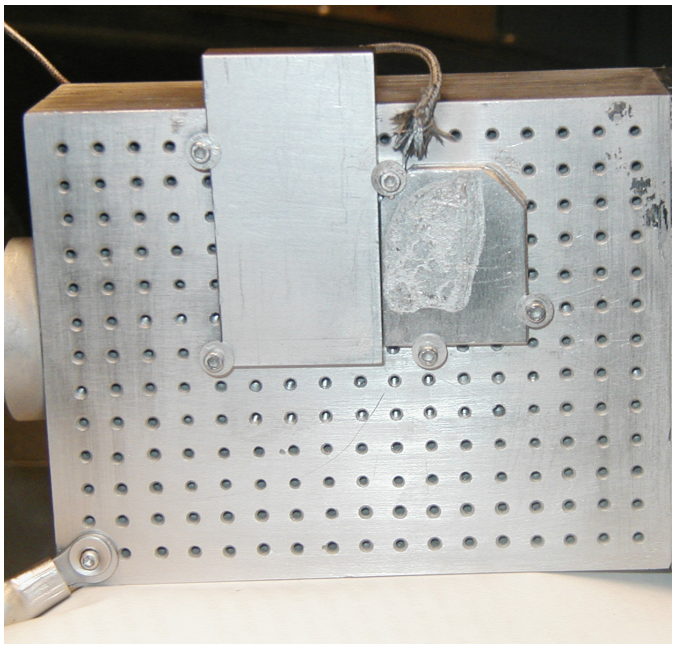

Figure 9. TZM Substrate mounted on a water-cooled holder. Additionally one can see a piece of Mo-coated silicon (right of the large TZM piece) and a thermocouple wire (top center)

A number of conclusions could be drawn from these tests:

- Prolonged operation of the source is possible but parts of the chamber, which is a ground and thus at anode potential, gets very warm (eventually to hot to touch!);
- A 10-sec arc pulse let the thermocouple rise by about 100 degrees. Successive pulsing with long pulses lead to surface temperatures exceed 200°C
- the adhesion is greatly improved when the substrate holder is negatively biased
- For films thicker than 1 μm, adhesion is not satisfactory unless negative bias is used AND substrate surface has been thoroughly cleaned (see below).

VI. Adhesion

The adhesion was tested using the cheap industry standard tape test: Scotch "Magic Tape 440" was pressed against the coating and the pulled off. The "tape test" is considered passed only if no film sticked to the tape upon removal from the substrate. To make sure film adhesion is good, several negative bias voltages were tested. Bias was applied from 400 V, 5 A power supply. It was found that bias of –200 V was required, especially when the initial interface between the TZM substrate and the Mo film was formed. The effect of bias can be understood in terms of ion acceleration and sputtering. The average charge state of vacuum-arc-produced molybdenum is 3.1, and therefore a bias voltage of say –100 V will cause an average kinetic energy of 300 eV plus the initial kinetic energy the ion had before acceleration in the sheath. Details of these energetics are discussed in a recent publication [Anders, 2002 #1871]. Kinetic energies of order 500 eV or higher cause significant sputtering, thereby reducing the effective deposition rate.

VII. Deposition Rate

The deposition rate was determined by creating a step between a coated and uncoated area of the substrate. For example, a step can be produced by covering part of the substrate with the thin blocking piece such as a piece of silicon or glass or thin metal foil. The step was measured using a profilometer of the type Dektak 2A. We determined that, at substrate distance of 200 mm from the cathode surface, the deposition rate was 2.5 nm/s if the bias was –200 V. Lower bias gives higher effective rates since sputtering of Mo from the substrate is reduced; the rates are about 4 nm/s for –100 V and 5 nm/s for –50 Volt, which is close to the grounded case, i.e., maximum possible rate. These values are valid for an arc current of 70 A.

The source in operation is shown in Figure 10, and the cathode erosion is shown in Figure 11.

Figure 10: Arc source with Mo cathode in operation. A digital video clip is available.

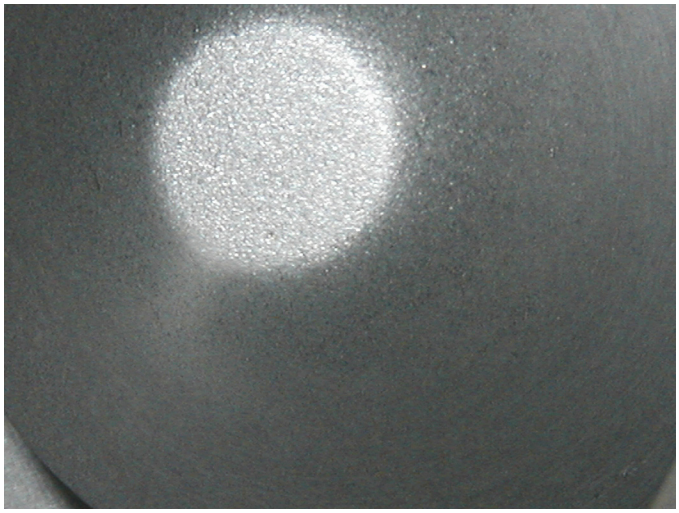

Figure 11: Tip of the conical molybdenum cathode after prolonged arcing. One can see the rough erosion structures caused by cathode spots. The area appearing bright is about 10 mm in diameter.

VIII. Deposition Parameters for Actual Beam Stopper Coating

The actual beam stopper was mounted on the water-cooled holder as shown in Figure 10.

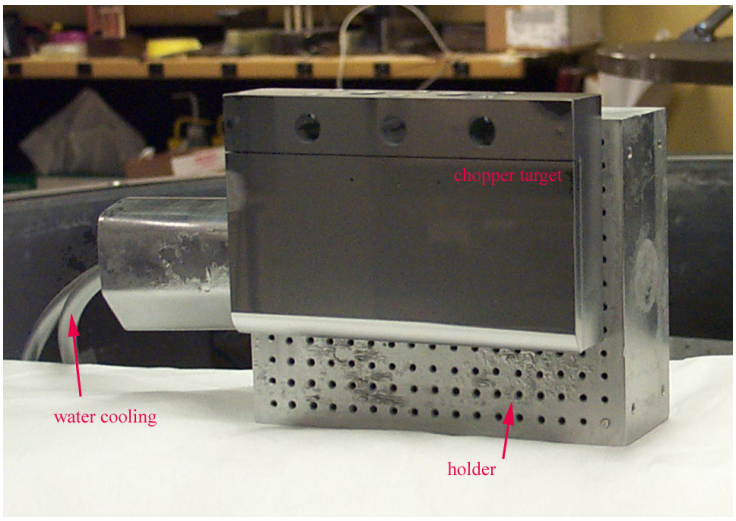

Figure 10. Beam stopper mounted on water-cooled holder (The defect near the left edge is an area of greater surface roughness due to imperfect machining, however, this should not affect the performance of the beam stopper). As expected, the defect was also visible after coating.

The holder was tilted by about 15° to make sure that the curved area at the lower edge obtains a maximum of deposition – this is where the ion beam has the highest intensity. The tilted holder is shown in Figure 11.

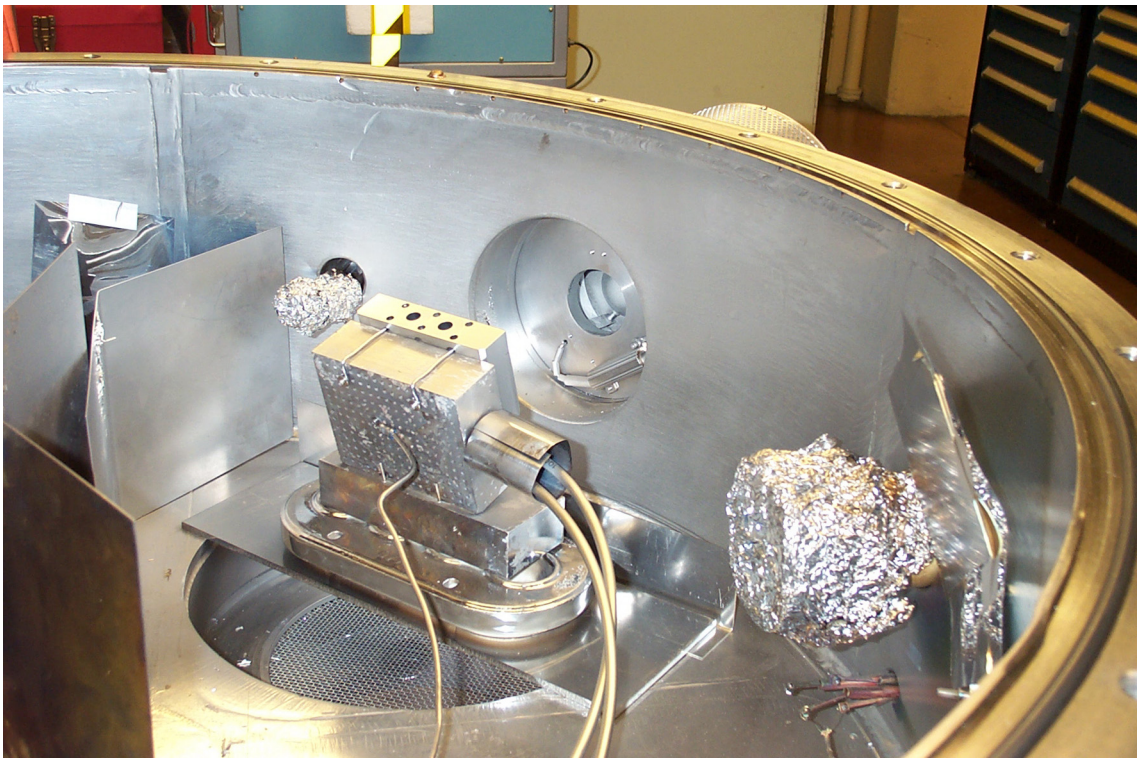

Figure 11: Vacuum chamber with tilted substrate holder.

The deposition was carried out after having the beam stopper "UHV cleaned" at LBNL's plating shop. The cleaning procedure is described in appendix A. The source was operated at a duty cycle of 33% with 10 s ON followed by 20 s OFF followed by 10 s ON etc. The duty cycle was a compromise between high average deposition rate and heating of the components, especially the chamber lit. The arc current was 70 A, and the vacuum base pressure in the high 10^{-6} Torr in the OFF periodes and in the 10^{-5} Torr range during the ON time. The first 400 s of arc operation (i.e., 1200 s total) were done with applied bias of –200 V,

corresponding to deposition rate of 2.5 nm/s (when the arc in ON). The coating thickness at this point was 1000 nm = 1 μm. Since the interface formation is by far completed at this point, bias can be reduced. We chose –100 V for the next 630 s or arc operation (giving about 2.5 μm thickness), followed by –50 V for 1000 s of arc operation, giving about 5 μm. The total deposition time (arc ON time was 2030 s, corresponding to about 34 minutes). The total approximate thickness is about 8.5 μm, giving us a wide safety margin for operational performance (see simulation section III, page 2).

Appendix A: Cleaning procedure for TZM (copied from LBNL memo 947)

1. 909 cleaner (mild caustic soap) for 15 minutes.
2. Rinse in de-ionized (DI) water.
3. HF acid for two minutes.
4. Tap water rinse.
5. Electropolish for two minutes:
6. volts 128°F
7. HCl 50% Dip
8. 909 cleaner.
9. DI water rinse.
10. DI water rinse.
11. Hot water, 160°F rinse
12. Nitrogen Airdry